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RELATIVISTIC CALCULATIONS AND MEASUREMENTS OF ENERGIES
AUGER RATES AND LIFETIMES(U) OREGON UNIV EUGENE DEPT OF
PHYSICS B CRASEMANN ET AL. DEC 82 AFOSR-TR-83-0550

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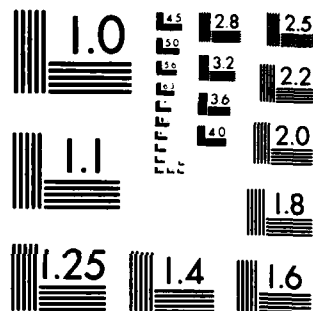
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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER AFOSR-TR- 83-0550	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) RELATIVISTIC CALCULATIONS AND MEASUREMENTS OF ENERGIES, AUGER RATES, AND LIFETIMES		5. TYPE OF REPORT & PERIOD COVERED ANNUAL SCIENTIFIC REPORT
7. AUTHOR(s) B. CRASEMANN AND M. H. CHEN		6. PERFORMING ORG. REPORT NUMBER
9. PERFORMING ORGANIZATION NAME AND ADDRESS DEPARTMENT OF PHYSICS UNIVERSITY OF OREGON EUGENE, OREGON 97403		8. CONTRACT OR GRANT NUMBER(s) AFOSR 79-0026
11. CONTROLLING OFFICE NAME AND ADDRESS AFOSR (NP) BOLLING AFB, DC 20332		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F 230/A4
13. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		12. REPORT DATE DECEMBER 1982
15. DISTRIBUTION STATEMENT (of this Report) APPROVED FOR PUBLIC RELEASE; DISTRIBUTION UNLIMITED		13. NUMBER OF PAGES 18
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		15. SECURITY CLASS. (of this report) UNCLASSIFIED
19. SUPPLEMENTARY NOTES		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) ATOMIC INNER-SHELL PROCESSES X-RAY EMISSION AUGER TRANSITIONS SYNCHROTRON RADIATION INNER-SHELL IONIZATION BY PROTONS BREIT INTERACTION DIRAC-HARTREE-SLATER COMPUTATIONS RESONANT AUGER RAM EFFECT		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Substantial progress has been made with <u>ab initio</u> relativistic computations of atomic inner-shell energy levels and properties of few-electron ions. A relativistic calculation of inner-shell ionization by slow protons has been very successful. Threshold-excitation experiments with hard synchrotron radiation have been extended to explore post-collision interaction and the resonant Auger Raman effect which link the atomic excitation and deexcitation processes.		

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Department of Physics
and
Chemical Physics Institute



Relativistic Calculations and Measurements
of Energies, Auger Rates, and Lifetimes

U. S. Air Force Office of Scientific Research
Grant No. AFOSR-79-0026

ANNUAL REPORT

1 December 1981 - 30 November 1982

1. Introduction

The main emphasis in this research program is upon state-of-the-art computations, including relativistic and quantum-electrodynamic effects, of atomic energy levels and of x-ray and Auger transitions in atoms with one or several deep vacancies -- or in their counterparts, viz., few-electron heavy ions. The theoretical work is complemented by experimental studies of inner-shell threshold-excitation and resonance phenomena using gas-phase electron spectrometry with synchrotron radiation. In an additional, new effort, relativistic self-consistent-field wave functions have been used in improved calculations of atomic inner-shell ionization by charged particles.

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MATTHEW J. KENTER
Chief, Technical Information Division

2. Status of the Research

a) Atomic energy levels

Access to the large computing facilities at the NASA Ames Research Center has enabled us to perform relativistic self-consistent-field computations of atomic inner-shell level energies, including quantum-electrodynamic corrections. At the same time, highly precise experimental level energies have become available from modern precision measurements of x-ray energies (notably at the National Bureau of Standards and at the Kernforschungsanlage Jülich in West Germany), and from absorption measurements with synchrotron radiation, interpreted in terms of theoretical edge shapes. Systematic comparison of calculated and measured atomic level energies has led to significant new insights. We have been particularly concerned with the effect of interaction with Auger continua, the effect of screening of the self energy, and the effect of the nuclear charge distribution on inner-shell energies, the effect of the Breit interaction on energy splittings between multiple-hole states, and configuration-interaction effects in core-level spectra.

A systematic discrepancy between measured and calculated 2s-level energies has been noted throughout the periodic table. Predicted 2s levels lie several electron volts above measured energies (~ 4 eV for $30 \leq Z \leq 47$), while the energies of adjoining levels can be computed much more closely. A comparable problem appears to exist for the 3s levels. Configuration interaction

with nearby bound states does not explain the difficulty. We were able to show, however, that the interaction between the bound atomic electrons and the radiationless continua of Auger and Coster-Kronig states does account for the observed depression of atomic 2s levels up to $Z=50$ [Phys. Rev. A 24, 1158 (1981)]. This interaction is the radiationless analog of the electron self energy. Above $Z=50$, a residual discrepancy remains that appears to grow with atomic number; this remaining discrepancy may well be due to a quantum-electrodynamic effect but has not yet been explained.

The interaction of bound electrons with the radiative continuum has been known for a considerable time; it gives rise to the self energy which together with the vacuum polarization comprises the Lamb shift. The electron self energy becomes an important correction to inner level energies of heavy atoms (Fig. 1). A difficulty arises from the fact that the effect of screening on the self energy is not yet fully understood. Only for the K shell have screened self-energy shifts been calculated to date. We have had good success, however, with an effective-charge approach to estimate screening of the self energy in the L shells. Using this approach and a Fermi distribution of the nuclear charge and the frequency-dependent form of the Breit interaction, we have made a new computation of the innermost level energies of heavy atoms [At. Data Nucl. Data Tables 26, 561 (1981)]. These results have greatly reduced discrepancies with

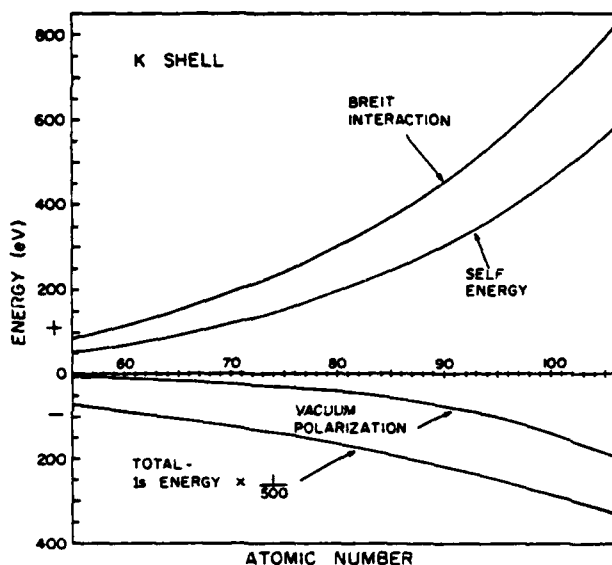


Fig. 1. Shifts produced in the 1-s level energy of heavy atoms by the Briet interaction, self energy, and vacuum polarization. From M. H. Chen et al., At. Data Nucl. Data Tables 26, 561 (1981).

the most precise recent experiments [cf. E. G. Kessler, Jr., R. D. Deslattes, D. Girard, W. Schwitz, L. Jacobs, O. Renner, preprint and abstract of paper presented at the X-82 Conference on X-Ray and Atomic Inner-Shell Physics; G. Barreau, H. G. Borner, T. von Egidy, and R. W. Hoff, preprint of paper submitted to Z. Physik, see Fig. 2].

Extensive computations of L x-ray satellite energies, produced as a byproduct of our other energy calculations, have now been published [At. Data Nucl. Data Tables 26, 383 (1981)]. These tables include the energies of all L x-ray satellites that arise from electric dipole and quadrupole transitions in the presence of one spectator hole in the M or N shell, in 11 elements with $65 \leq Z \leq 95$. The computations are relativistic and include quantum-electrodynamic corrections. In subsidiary

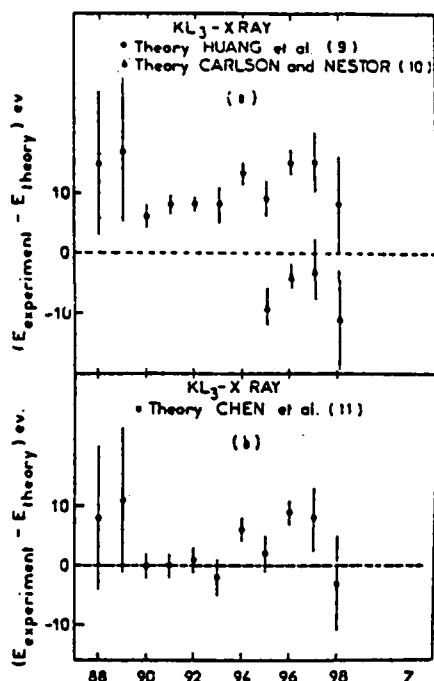


Fig. 2. Comparison between calculated K x-ray energies and measurements by G. Barreau et al. in the Institut Laue-Langevin, Grenoble. The lower panel refers to our most recent work.

tables, we list the Coulomb and Breit interaction energies for double-hole states and the relative x-ray intensities of double-hole multiplet states.

The tightly bound electrons near the nucleus move in a strong field at an appreciable fraction of the speed of light, particularly in heavy atoms. Atomic inner shells therefore provide a proving ground for calculations of relativistic effects. In a particularly elegant formulation, the Breit operator comprises all of the two-electron effects outside the instantaneous Coulomb interaction (to lowest order in the fine-structure constant), viz., the spin-other-orbit, spin-spin, and orbit-orbit interactions. The Breit operator was refined further

in recent years to take account of its dependence on the frequency of the virtual photon. The operator plays such a central role in relativistic atomic theory that detailed experimental tests are relevant. In most contexts, the Breit interaction manifests itself quite subtly, and is not easily evaluated with precision (Fig. 1). A notable exception is found in states of several atomic inner-shell holes: due to partial cancellation of the instantaneous Coulomb part of the Hamiltonian, the multiplet splitting between different couplings of deep holes is grossly influenced by the Breit energy, which even causes the level order to be reversed in some cases. The most striking measurable Breit effect probably is the shift of so-called K x-ray hypersatellites. These x-ray lines arise from radiative transitions to a completely empty 1s shell in atoms from which both K electrons have been removed; a full 20% of the $K\alpha_1$ hypersatellite shift at high Z is due to the Breit energy! Our extensive computations of K x-ray hypersatellite shifts have recently been published [Phys. Rev. A 25, 391 (1982)]. These results are being compared with new measurements [C.W.E. van Eijk, J. Wijnhorst, and M. A. Popelier, Phys. Rev. A 24, 854 (1981); Y. Isozumi, Ch. Briançon, and R.J. Walen, preprint of paper submitted to Phys. Rev. C].

We have started a collaboration with colleagues in the Universities of Linköping, Sweden, and Aarhus, Denmark, to work on a systematic review of configuration-interaction effects on

core-level energies in metals. A preliminary report on this work was presented at the X-82 Conference.

b) Properties of few-electron heavy ions

Radiative and radiationless transition probabilities have been computed relativistically for Li-, N-, F-, K-, and Co-like ions with atomic numbers up to $Z=92$ and with initial configurations consisting of a few closed inner shells with one deep-lying vacancy. Relativity was found to have a pronounced effect on the Auger rates of these highly ionized systems. Radiationless-transition rates were seen to increase with degree of ionization; the stripping of outer shells has a much smaller effect than creation of an empty inner shell of spectator holes. Fluorescence yields were found to increase with the degree of ionization. In the radiative decay of $1s2s^2$ Li-like ions, initial-state correlation boosts the fluorescence yield by six orders of magnitude at low and medium Z .

The properties of the $1s2p^2$ states of Li-like ions were calculated relativistically for $6 \leq Z \leq 30$, with particular attention to the effects of the Breit interaction on multiplet splitting and on the radiationless transitions through which these states decay. The full Breit interaction (magnetic and retardation terms) was included in calculating fine structure. Transition rates were computed in relativistic intermediate coupling with configuration interaction. Both the spin-orbit and magnetic

interactions were incorporated in the calculations. Results were compared with earlier calculations and with experiment, and the physics of the observed relativistic and configuration-interaction effects has been analyzed.

A substantial effort was devoted to computations of relativistic Auger and x-ray emission rates of $1s2s2p$ configurations of three-electron ions. Relativistic effects on the Auger rates were seen to be as large as two orders of magnitude; multiplet fluorescence yields were found to be substantially affected, and the Breit interaction was seen to reverse the fine-structure level order in some cases. These excited ions are of particular interest because of the metastability of the $4p$ states, which could make it possible to produce a population inversion with quite limited pumping power. In addition, such highly stripped atoms play an important role in astrophysical and plasma environments. Many of the present theoretical results are amenable to experimental test, and some related measurements already exist.

c) Inner-shell ionization by charged particles

We have used our Dirac-Hartree-Slater wave-function code to calculate Coulomb ionization of the L subshells of Au and U by slow protons, in the relativistic plane-wave Born approximation. Semiclassical corrections for binding-energy change and Coulomb deflection were applied. Considerable improvement was attained in these ab initio relativistic PWBA calculations with DHS wave

functions, as compared with earlier, classical, semiclassical, and PWBA results based on screened hydrogenic wave functions. The predicted subshell ionization cross sections agree well with experimental data. The effect of relativity tends to partially cancel the effects of binding and Coulomb deflection. The calculated energies at which minima occur in the L_1/L_2 and L_3/L_2 cross-section ratios also agree well with observation. Predicted total L x-ray cross sections agree extremely well with experiment. The $L\alpha/L\gamma$ and $L\alpha/L\beta$ x-ray emission rates for p+Au agree with measurements if L fluorescence and Coster-Kronig yields are adjusted in accordance with recent relativistic theory. This work has most recently been extended to the M shell, with considerable success.

d) Inner-shell threshold excitation with synchrotron radiation

Atomic inner-shell hole states have very short lifetimes -- typically, a few femtoseconds. Consequently, the levels are several electron volts broad. The large width of these short-lived hole states invites exploration of threshold excitation phenomena with synchrotron radiation that has been monochromatized to a band width far less than one electron volt. This has led to the exploration of energetic resonance phenomena, such as the Auger resonant Raman effect which our group discovered [Phys. Rev. Lett. 45, 1937 (1980)]. Here, the excitation and deexcitation of a deep hole state are no longer

separable but are described by a single, second-order matrix element. The electrons (or, in the analogous x-ray resonant Raman effect, the photons) emitted in the deexcitation produce a line that is narrower than the natural linewidth of the normal radiation, and the cross section is resonantly enhanced at threshold. Applications of these phenomena are still in a speculative stage; they are likely to lead to energy measurements of exceptional precision, new techniques of sensing the extraatomic environment, and new ways of probing the time development of atomic deexcitation.

We have built a rather sophisticated gas-phase electron spectrometer that is operated on-line at the Stanford Synchrotron Radiation Laboratory. During the past year, we were granted one block of 22 eight-hour shifts of beam time, beginning 20 January 1982, on the 8-pole wiggler line. The wiggler yields approximately 60 times the flux of ordinary bending-magnet lines; this higher intensity was necessary for our experiments. We obtained excellent threshold-excitation data for three transitions in argon and xenon. The purpose of the experiments was (i) to obtain more precise data on the Auger resonant Raman effect, in several different transitions, and (ii) to make accurate measurements of the post-collision interaction shift of Auger and Coster-Kronig lines, in order to test the semi-classical Niehaus theory and measure vacancy lifetimes. The data

are still being analyzed; a preliminary report was given at the X-82 Conference.

e) International Conference on X-Ray and Atomic Inner-Shell Physics

The International Conference on X-Ray and Atomic Inner-Shell Physics ("X-82") was held on the University of Oregon campus on 23-27 August 1982. Our group has been deeply involved in preparations for the conference that brought together 222 scientists from 26 different countries. The conference was a sequel to similar meetings held in Freiburg in 1976, Sendai in 1978, and Stirling (Scotland) in 1980. The program included 56 invited papers and 164 contributed papers on atomic inner-shell transitions, x-ray scattering and bremsstrahlung, and on the interface between x-ray physics and materials science, astrophysics and nuclear physics, the design of short-wavelength lasers, coherent generation in periodic structures, and controlled thermonuclear fusion. The five-day conference was sponsored by the International Union of Pure and Applied Physics, the American Physical Society, the University of Oregon, and by the U. S. Department of Energy, the Army Research Office, and the National Science Foundation. The Proceedings will be published in December, 1982, as a volume in the American Institute of Physics Conference Proceedings Series.

3. Publications

Published Papers

1. F. Parente, M. H. Chen, B. Crasemann, and H. Mark: "L X-Ray Satellite Energies." At. Data Nucl. Data Tables 26, 383 (1981). [Actual publication date March, 1982].
2. M. H. Chen, B. Crasemann, M. Aoyagi, K.-N. Huang, and H. Mark: "Theoretical Atomic Inner-Shell Energy Levels, $70 \leq Z \leq 106$." At. Data Nucl. Data Tables 26, 561 (1981). [Actual publication date March, 1982].
3. B. Crasemann: "Relativity: X-Ray and Auger Transitions." In Inner-Shell and X-Ray Physics of Atoms and Solids, edited by D. F. Fabian, H. Kleinpoppen, and L. M. Watson (Plenum, New York, 1981), p. 97.
4. M. H. Chen, B. Crasemann, and H. Mark: "Effect of the Breit Interaction on K X-Ray Hypersatellite Spectra." Phys. Rev. A 25, 391 (1982).
5. M. H. Chen, B. Crasemann, and H. Mark: "Atomic L-Shell Ionization by Protons: Dirac-Hartree-Slater Calculation of Cross Sections." Phys. Rev. A 26, 1243 (1982).
6. M. H. Chen, B. Crasemann, and H. Mark: "Effects of Relativity on Multiplet Splitting and Decay Rates of the $1s2p^2$ Configuration of Li-Like Ions." Phys. Rev. A 26, 1441 (1982).

In Press and Submitted

7. M. H. Chen, B. Crasemann, and H. Mark: "Deexcitation of Light Li-Like Ions in the $1s2s2p$ State." Phys. Rev. A, in press.
8. M. H. Chen, B. Crasemann, and H. Mark: "K-MM Auger-Intensity Peaks from Double-Hole Energy-Level Crossings." Phys. Rev. A, Rapid Communications, in press.
9. M. H. Chen, B. Crasemann, and H. Mark: "Radiationless Transitions to Atomic $M_{1,2,3}$ Shells: Relativistic Theory." Submitted to Physical Review A.
10. M. H. Chen, B. Crasemann, and H. Mark: "Relativistic Calculation of Atomic M-Shell Ionization by Protons." Submitted to Physical Review A.
11. M. H. Chen: "Relativistic Effects in Atomic Inner-Shell Transitions." In Proceedings of the X-82 International Conference on X-Ray and Atomic Inner-Shell Physics, edited by B. Crasemann (American Institute of Physics Conference Proceedings Series, New York, in press).
12. G. E. Ice, G. S. Brown, G. B. Armen, M. H. Chen, B. Crasemann, J. Levin, and D. Mitchell: "Atomic Inner-Shell Threshold Excitation with Synchrotron Radiation." In Proceedings of the X-82 International Conference on X-Ray and Atomic Inner-Shell Physics, edited by B. Crasemann (American Institute of Physics Conference Proceedings Series, New York, in press).

13. B. Crasemann: "Atomic Inner-Shell Transitions---Theory and the Need for Experiments." In Proceedings of the 7th Conference on the Application of Accelerators in Research and Industry, to be published.

Abstracts

14. B. Crasemann: "Relativistic Effects on Inner-Shell X-Ray and Auger Transitions." Bull. Am. Phys. Soc. 27, 41 (1982).
15. M. H. Chen, B. Crasemann, and H. Mark: "Breit Interaction Effect on Multiplet Splitting and Hypersatellite Shifts." Bull. Am. Phys. Soc. 27, 515 (1982).
16. M. H. Chen, B. Crasemann, and H. Mark: "Relativistic Computations of L-Shell Ionization by Slow Protons." Program and Abstracts, X-82 International Conference on X-Ray and Atomic Inner-Shell Physics (unpublished), Vol. I, p. 116.
17. N. Mårtensson, M. H. Chen, B. Crasemann, and B. Johansson: "Systematics of CI Effects in Core-Level Spectra---A Comparative Study of Experimental and Δ SCF Core Ionization Energies." Program and Abstracts, X-82 International Conference on X-Ray and Atomic Inner-Shell Physics (unpublished), Vol. II., p. 225.
18. B. Crasemann, M. H. Chen, G. B. Armen, J. Levin, G. E. Ice, and G. S. Brown: "Threshold Excitation Effects in Atomic Inner-Shell Auger Spectra." In Ninth Annual Stanford Synchrotron Radiation Laboratory Users Group Meeting,

October 21-22, 1982, edited by I. Lindau and M. Klein. SSRL
Report No. 82-04 (unpublished), p. 1.

4. Professional Personnel

Bernd Crasemann, Professor of Physics, Principal Investigator.

Mau Hsiung Chen, Senior Research Associate, Co-Principal
Investigator.

G. Bradley Armen, Research Assistant.

Kh Rezaul Karim, Research Assistant.

Jon Levin, Research Assistant.

Quoc Oai Vu, Student Programmer (to 1 September 1982).

Off-Campus Collaborators:

George S. Brown, Stanford Synchrotron Radiation Laboratory.

Gene E. Ice, Oak Ridge National Laboratory.

Hans Mark, National Aeronautics and Space Administration.

5. Interactions

a) Spoken Papers Presented at Meetings, Conferences, etc.

1. B. Crasemann: "Relativistic Effects on Inner-Shell X-Ray and Auger Transitions." Invited talk at the 1982 Joint Annual Meeting of the American Physical Society and the American Association of Physics Teachers, San Francisco, 25-28 January 1982 (see Sec. 3, Item 14).
2. M. H. Chen, B. Crasemann, and H. Mark: "Breit Interaction Effect on Multiplet Splitting and Hypersatellite Shifts."

Contributed paper at the Spring Meeting of the American Physical Society, Washington, D.C., 26-29 April 1982 (see Sec. 3, Item 15).

3. M. H. Chen: "Relativistic Effects in Atomic Inner-Shell Transitions." Invited talk at the X-82 International Conference on X-Ray and Atomic Inner-Shell Physics, Eugene, 23-27 August 1982 (see Sec. 3, Item 11).
4. B. Crasemann: "Threshold Excitation Effects in Atomic Inner-Shell Auger Spectra." Invited talk at the Ninth Annual Stanford Synchrotron Radiation Users Group Meeting, 21-22 October, 1982 (see Sec. 3, Item 18).
5. B. Crasemann: "Atomic Inner-Shell Transitions---Theory and the Need for Experiments." Invited talk at the 7th Conference on the Application of Accelerators in Research and Industry, Denton, Texas, 8-10 November 1982 (see Sec. 3, Item 13).
6. B. Crasemann: "Highlights of the X-82 International Conference on X-Ray and Atomic Inner-Shell Physics." Invited talk at the 7th Conference on the Application of Accelerators in Research and Industry, Denton, Texas, 8-10 November 1982.
7. B. Crasemann: "Efectos Relativísticos y de QED Sobre las Transiciones Rayos-X y Auger Entre las Capas Internas Atómicas." Invited lecture at the Physics Department,

Universidad de Santiago de Chile, 23 March 1982 (in Spanish).

8. B. Crasemann: "Synchrotron Radiation as a Probe of Atomic Inner Shells." Colloquium lecture, Physics Department, New Mexico State University, Las Cruces, 11 November 1982.

b) Consultative and Advisory Functions

1. Stanford Synchrotron Radiation Laboratory Proposal Review Panel, meetings 14-16 January 1982 and 17-19 June 1982.
2. X-82 Conference Organizing Committee: Local Committee Meeting 6 March 1982, Eugene; Program Committee Meeting 30 April 1982, Washington, D.C.; Conference, 23-27 August 1982, Eugene.
3. Executive Committee, Division of Electron and Atomic Physics, American Physical Society, meetings 27 April 1982, Washington D.C., and 3 November 1982, Philadelphia, Pa.
4. Ad hoc Panel for the Evaluation of the Atomic Physics Research Program at Brookhaven National Laboratory, Upton, N.Y., 24-25 May 1982.
5. Ad hoc Committee for the Review of the Doctoral Program in the Natural Sciences in the University of Nevada, Reno, 6-8 October 1982.
6. Council, American Physical Society, Philadelphia, Pa., 5-6 November 1982.
7. Editorial Board, Atomic Data and Nuclear Data Tables, Washington D.C. 28 April 1982.